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Intercrystalline heat conductivity in the presence of planar defects

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Abstract. The heat flux between two solids is calculated, taking into account the impurity monolayer separating the crystals. It is found that the weak impurity–crystal interaction results in a resonance-type frequency dependence of the phonon energy transmission coefficient. The calculation enabled a description of the low-temperature heat transfer anomalies recently measured in point contacts to be given.

1. Introduction

The phonon transport through the interface between two crystals is of particular importance for basic research [1, 2]. The generalized dynamic model of the intercrystalline boundary can be described as a two-dimensional (planar) defect of the crystal [3, 4]. Such a defect may be a packing defect, or an impurity layer which is monotonically thick.

The investigation of solid–solid phonon transport is stimulated by point contact production, which enables us to study the ballistic heat conductivity [5, 6]. Ballistic transport in point contacts is by almost non-interacting groups of carriers coming from the opposite edges. A highly non-equilibrium state of the phonon system can be realized in conducting and insulating point contacts if the contact diameter d is small compared with the phonon relaxation length l_{ph} in the bulk, but it is much larger than the phonon characteristic wavelength. Near the junction there can be no equilibrium temperature, and the thermal flux cannot be calculated in terms of the traditional thermodynamics of non-equilibrium processes.

The problem of ballistic phonon transport is important because the inelastic phonon–phonon scattering length $l_{ph\text{-}ph}$, which is associated with anharmonicity of crystal lattice vibrations, rapidly increases with decreasing temperature T so that, at $T < 10$ K, contacts of diameter $d < 10^3$ nm can be regarded as point contacts. Heat conduction between contacting solids is in many cases realized through a number of parallel point contacts. At low temperatures, heat removal can also be ballistic in microelectronics, with the device size now being as small as almost 10^2 nm. Also, the heat conduction of point contacts can be used to study the surface properties of solids. The reason is that the thermal resistance of a point contact is much higher than that of the edges; therefore the heat flux depends on the state of a small region immediately surrounding the point contact orifice and having a size of the order of the point contact diameter.

The experimental data presented in [5–8] show that, in dielectric point contacts manufactured by pressing, at $4\ \mathrm{K} < T < 30$ K, the temperature dependence of the heat

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conductivity differs greatly from the results of the acoustic mismatch theory [9]. At NaCl–NaCl [5, 6, 8], KBr–KBr and KBr–Cu [7] point contacts an anomalous increase in the point contact thermal conductivity at low temperatures has been revealed. This feature could be due to the properties of the real interface of the media.

In the present paper, such anomalies are interpreted as resulting from the presence of a planar defect at the point contact boundary. The intercrystalline phonon transport is mathematically modelled on the basis of the 'capillary' theory [10–13] of phonon transmission through impurity layers which are monatomically thick. Study of the capillary effects in terms of the macroscopic theory of elasticity is based on consideration of phenomena similar to the Laplace excessive pressure arising because of a two-dimensional defect in the crystal [3, 4].

2. Ballistic phonon transport at a point contact

We shall model the point contact as an orifice in a plane screen reflecting the phonons. In reality the surface of the screen is that of the vacuum gap forming the point contact (figure 1).

For further calculations we assume that both edges of the contact are made of the same dielectric and consider phonon reflection and refraction by the point contact boundary.

Near the point contact, ballistic spreading of phonons occurs, and therefore the heat flow through a single point contact is not associated with an appreciable change in temperature near the point contact (this has been considered in detail in [8]). Therefore heat is transferred by groups of phonons whose distribution functions depend on the massive edge temperatures (T and T_0) [8, 14, 15].

The temperature difference $\Delta T = T - T_0$ may be made arbitrarily large. This can be represented as a realization of the 'point contact Kapitza temperature discontinuity' arising from phonon scattering by the vacuum gap boundaries.

Then the heat flux $\dot{Q}_{\mathrm{B}}$ from the contact edge having the temperature T to the edge kept at the constant temperature T_0 can be written as [8]

$$\dot{Q}_{\mathrm{B}} = \frac{\hbar A}{2(2\pi)^3} \sum_j \int \mathrm{d}\boldsymbol{k}\, \omega_j(\boldsymbol{k}) |s_z^j| D^j(\boldsymbol{k}) \left[N\left(\frac{\omega_j}{T}\right) - N\left(\frac{\omega_j}{T_0}\right) \right]. \tag{1}$$

Here $D(\boldsymbol{k})$ is the coefficient of phonon energy transmission through the point contact boundary and $\boldsymbol{s} = \partial\omega/\partial\boldsymbol{k}$; $N(x) = 1/[\exp(\hbar x) - 1]$ is Planck's distribution function; the z axis is normal to the point contact boundary, A is the contact area, and j is the phonon mode; ω and $\boldsymbol{k}$ are the phonon frequency and wavevector.

Assuming that $D(\omega) =$ constant at low temperatures T, $T_0 \ll \Theta_{\mathrm{D}}$ (here Θ_{D} is the Debye temperature), we obtain the estimate

$$\dot{Q}_{\mathrm{B}} = A D s^{-2} \hbar^{-3} (T^4 - T_0^4). \tag{2}$$

The low-temperature dependence of the heat flux is similar to that obtained by Little [9] who dealt with the thermal resistance due to acoustic mismatch between the two media.

If it is assumed that the presence of the point contact does not essentially alter the phonon dispersion law, then calculation of the heat flux amounts to finding the transmission coefficient D.

3. Resonance phonon transport through the planar defect

In the presence of an intermediate boundary layer weakly bonded with the contact edges, there can be a resonance heat transfer mechanism. Such a mechanism can be described in terms of both the discrete lattice dynamics [16] and the theory of elasticity, if the capillary effects are taken into consideration [17].

As was shown in [11] and [18] to describe the dynamics of a weakly bonded impurity layer, it is appropriate to introduce an additional independent dynamic variable (an internal degree of freedom of the layer). This variable plays the role of the elastic displacement $u^{(s)}$ of the layer, which in the general case is different from the displacements of the surfaces of the neighbouring media.

To derive the dynamic boundary conditions at the interface of a two-dimensional lattice defect, in the bulk equations of motion it is necessary to vary the free energy E of the system. E is the sum of the bulk contribution E_v and surface contribution E_s. The variation in the bulk energy (taking account of the equation of motion $\rho u_i = \partial\sigma_{ik}/\partial x_k$) is equal to

$$\delta E_v = \int \mathrm{d}S\,(\sigma_{in}^{(1)}\,\delta u_i^{(1)} - \sigma_{in}^{(2)}\,\delta u_i^{(2)}) \tag{3}$$

where the integral is taken over the undeformed boundary of two solid bodies, $\sigma_{in}^{(1,2)} = C_{iklm}^{(1,2)} u_{lm}$ is the stress tensor, u_i and u_{ik} are the elastic displacement vector and the deformation tensor, the $C_{iklm}^{(1,2)}$ are the bulk moduli of elasticity of the media in contact, $\sigma_{in} = \sigma_{ik} n_k$, and n_i is a unit vector normal to the surface, directed from medium 1 to medium 2.

The variation in the surface energy takes the form

$$\delta E_s = \int \mathrm{d}S\,\delta\alpha \tag{4}$$

where $\delta\alpha$ is the variation in the free-surface-energy density, for which we have the following thermodynamic identity:

$$\delta\alpha = g_{\mu j}\,\delta u_{\mu j}^{(s)} + \sigma_{in}^{(2)}(\delta u_i^{(2)} - \delta u_i^{(s)}) + \sigma_{in}^{(1)}(\delta u_i^{(s)} - \delta u_i^{(1)}) \tag{5}$$

where $g_{\alpha\beta}$ is the surface tension tensor; the Latin subscripts take the values 1, 2, 3, and the Greek subscripts 1, 2 and number the axes of coordinates in the plane tangential to the boundary. In equation (5), $u_i^{(s)}$ is the vector displacement of the boundary between the two solid media, which, owing to the presence of a discontinuity of the displacements of the media in contact ($\Delta_i = u_i^{(2)} - u_i^{(1)} \neq 0$), on such a boundary is different from $u_i^{(2)}$ and $u_i^{(1)}$.

Equating to zero the variation in the total free energy (taking account of the surface kinetic energy), we obtain the following effective boundary conditions for the surface stresses $\sigma_{ik}^{(1,2)}$ and displacements $u_i^{(1,2)}$ at the planar defect:

$$\sigma_{in}^{(1)} - \sigma_{in}^{(2)} = -\rho_s \ddot{u}_i^{(s)} + g_{\alpha,\beta}^{(0)} \nabla_\alpha \cdot \nabla_\beta u_i^{(s)} + \delta_{i,\beta} h_{i\beta j\delta} \nabla_\alpha u_{j\delta}^{(s)} \tag{6}$$

$$u_i^{(1)} - u_i^{(s)} = -b_{ik}\sigma_{kn}^{(1)} - C_{ik}\sigma_{kn}^{(2)} \tag{7}$$

$$u_i^{(s)} - u_i^{(2)} = -b_{ik}\sigma_{kn}^{(2)} - C_{ik}\sigma_{kn}^{(1)}. \tag{8}$$

A plane defect is determined by the capillary parameters ρ_s, $g^0_{\alpha\beta}$, $h_{\alpha\beta\gamma\delta}$, b_{ik} and c_{ik}, which characterize the excessive surface mass, the residual surface stress, the surface moduli of elasticity, and the force constants of the plane defect with respect to tangential and normal strains, respectively.

Let us consider for definiteness a shear wave incident at an angle θ to the normal z and polarized perpendicular to the plane XOZ. Considering a plane defect between two similar media we shall write $\omega^2 = k^2 C_{44}/\rho$ where ρ is the crystal density and C_{44} is the modulus of elasticity. We shall assume that the system (6)–(8) has a solution in the form

$$\begin{aligned} u_y^{(1)} &= u_0[\exp(ik\cos\theta z) + r\exp(-ik\cos\theta z)]\exp(ik\sin\theta x - i\omega t) \\ u_y^{(2)} &= u_0 d\exp(ik\cos\theta z + ik\sin\theta x - i\omega t) \\ u^{(s)} &= u_s^{(0)}\exp(-ikx\sin\theta - i\omega t). \end{aligned} \tag{9}$$

Let us write the expression for the reflection amplitude coefficient r_a and the transmission amplitude coefficient d_a of a plane defect between two similar media [11, 12] as

$$r_a = A/E \qquad d_a = B/E \qquad u_s = u_0 C/E \tag{10}$$

where

$$A = C_{44}^2 k\cos^2\theta(c_2 + b_2) - \tfrac{1}{2}[(\rho_s/\rho)kC_{44} - (g_1 + h_{66})k\sin^2\theta][1 + C_{44}^2 k^2\cos^2\theta(b_2^2 - c_2^2)] \tag{11}$$

$$B = C_{44}i\cos\theta + C_{44}c_2 i\cos\theta[(\rho_s/\rho)k^2 C_{44} - (g + C_{66})k^2\sin^2\theta] \tag{12}$$

$$C = C_{44}i\cos\theta + C_{44}^2 k\cos^2\theta(c_2 + b_2) \tag{13}$$

$$\begin{aligned} E = {} & C_{44}i\cos\theta + C_{44}^2 k\cos^2\theta(c_2 + b_2) + \tfrac{1}{2}[(\rho_s/\rho)kC_{44} - (g_1 + h_{66})k\sin^2\theta] \\ & \times [c_2^2 C_{44}^2 k^2\cos^2\theta + (1 - b_2 C_{44}ik\cos\theta)^2]. \end{aligned} \tag{14}$$

In equations (11)–(14) the following notation is introduced: $g_1 = g_{xx}$, $h_{66} = h_{xyxy}$, $b_2 = b_{yy}$ and $c_2 = c_{yy}$.

If the plane defect is formed by a layer of impurity atoms weakly bonded to the point contact edges, then the following conditions can be fulfilled: $\rho_s/b_2 \ll \rho C_{44}$ and $\omega \gg b_2^{-1}(C_{44}\rho)^{-1/2}$. Such a system of impurity atoms is characterized by weakly dispersed optical-type vibrations of low frequency

$$\omega \simeq \omega_0 = [2\rho_s^{-1}(b_2 - c_2)^{-1}]^{1/2}. \tag{15}$$

The amplitudes of impurity layer displacements are much larger than those of the contacting media surfaces:

$$u_s/u_0 \simeq (C_{44}\rho b_2/\rho_s)^{1/2} \gg 1. \tag{16}$$

Outside the resonance region we have $r_a \simeq 1$, $d_a \ll 1$, because of weak acoustic coupling between the edges. In the resonance range ($\omega \simeq \omega_0$), for non-slipping angles of incidence the wave is fully transmitted through the plane defect ($d_a \simeq 1$). A similar effect arises for sufficiently long waves if $\omega \ll \omega_0$.

If the transverse wave is incident on a plane defect, expressions (11)–(14) have the same form, up to the changes $C_{44} \rightarrow C_{11}$, $b_2 \rightarrow b_1$ and $c_2 \rightarrow c_1$.

The phonon energy transmission coefficient $D = D(\omega)$ is calculated from equation (10) as $D(\omega) = |d_a|^2$.

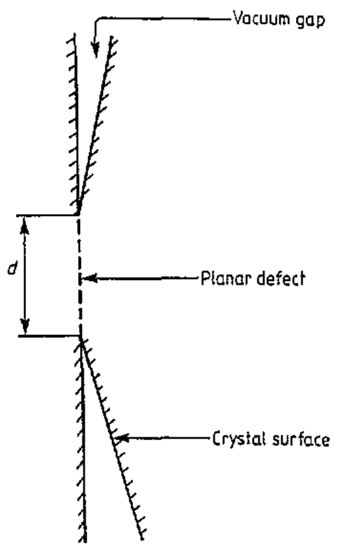

Figure 1. Diagram of a planar contact, formed by a short in the vacuum gap and containing the planar defect.

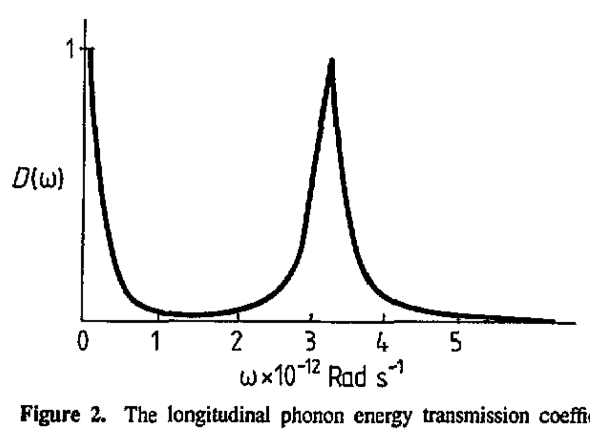

Figure 2. The longitudinal phonon energy transmission coefficient D as a function of the phonon frequency ω for the interlayer at a KBr–KBr point contact. The resonance frequency $\omega_0^{\mathrm{long}} = 3.0 \times 10^{12}$ rad s^{-1} is estimated from the experimental data [7, 8]. The thickness of the interlayer is chosen as 0.3 nm, and the density is 1 g cm^{-3}; then the corresponding elastic constant is $C_{33}^{\mathrm{int}} = 10^{-2} C_{33}^{\mathrm{KBr}}$.

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4. Heat flux calculation

Using the above equations (1) and (10), one can calculate the point contact heat flux $\dot{Q}_{\mathrm{B}}(T, T_0)$. Figure 3 shows the result of the calculation of heat flux due to the transmission of the longitudinal phonons at KBr–KBr point contacts. The function

$$F(T) = \dot{Q}_{\mathrm{B}}(T, T_0)/(T^4 - T_0^4) \tag{17}$$

is presented. Such presentation of data enables us to study the deviations from the simple T^4 law predicted by the acoustic mismatch model [9]. In this model it is assumed that $D(\omega) = \mathrm{constant}$. From the last assumption it follows that $F(T) = \mathrm{constant}$.

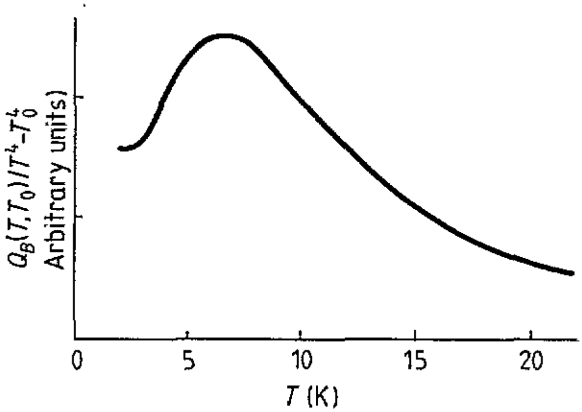

Figure 3. The reduced heat conductivity $F(T) = \dot{Q}_{\mathrm{B}}(T, T_0)/(T^4 - T_0^4)$ due to longitudinal phonon transmission through a KBr–KBr point contact ($T_0 = 2$ K; $\omega_0 = 3.0 \times 10^{12}$ rad s^{-1}).

The resonant frequency dependence of the transmission coefficient $D(\omega)$ significantly affects the temperature dependence of the thermal conductivity of the contact. In figure 3 the maximum of the function $F(T)$ (at $T_{\mathrm{max}} = 6$ K) is easily seen. The resonance frequency ω_0 is connected to T_{max} by the relation $\hbar\omega_0 = 3.89 T_{\mathrm{max}}$. The resonance frequency $\omega_0^{\mathrm{long}} = 3.0 \times 10^{12}$ rad s^{-1} is estimated from the experimental data [7, 8].

In figure 3 the beginning of the low-temperature increase at $T \simeq 2$ K arises because for phonons of the maximum wavelength the transmission coefficient D is equal to unity.

5. Discussion

The calculated extrema of the reduced heat conductivity $F(T)$ suggests that the intermediate impurity monolayer can have a significant effect on the phonon transport. Note that such low-frequency maxima can be induced by each of the phonon modes.

This type of situation goes beyond solid–solid contacts. It is known [1], for example, that heat transport between liquid helium and a solid in a certain temperature range can sharply increase from that predicted by the acoustic mismatch theory. One of the causes of this increase was considered in [16], where it is shown that the presence of a weakly bonded impurity monolayer gives rise to a resonance mechanism of heat transfer.

The transmission and reflection coefficients of a planar defect separating two different crystals can be analysed by the theory of capillary effects [12]. The above phenomena remain mainly the same in this case. Near the resonance frequency ω_0, the energy transmission factor is $D = |d|^2 Z_2/Z_1 \simeq 1$ (Z_1 and Z_2 are the acoustic impedances of the contacting media). While in the case of absence of the intermediate layer and strong acoustic mismatch of the media (e.g. for $Z_1 \gg Z_2$) the transmission factor D (and hence heat transfer) is very small, in the presence of the intermediate layer with the effective impedance $Z_0 = (Z_1/Z_2)^{1/2}$ at the resonance frequency there is complete transmission augmentation.

This effect is similar to the well known [19] complete transmission augmentation of optical systems including a macroscopic quarter-wave transmitting layer with the impedance equal to the geoemtric mean of the impedances of the contacting media. In the present case, unlike that in [19], the intermediate two-dimensional layer can have a thickness of the order of the atomic size.

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